

②

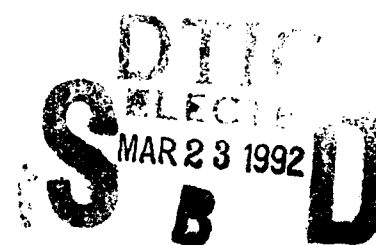


SINGLET DELTA OXYGEN PUMPING OF IODINE MONOFLUORIDE IN A SUPERSONIC FLOW

C. A. Helms
L. Hanko
G. P. Perram
G. Hager

January 1992

Final Report



APPROVED FOR PUBLIC RELEASE; DISTRIBUTION UNLIMITED.

92-07340



PHILLIPS LABORATORY
Directorate of Lasers and Imaging
AIR FORCE SYSTEMS COMMAND
KIRTLAND AIR FORCE BASE, NM 87117-6008

92 3 23 069

This final report was prepared by the Phillips Laboratory, Kirtland Air Force Base, New Mexico, under Job Order 31511Z22. The Laboratory Project Officer-in-Charge was Charles A. Helms (LDB).

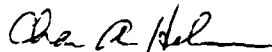
When Government drawings, specifications, or other data are used for any purpose other than in connection with a definitely Government-related procurement, the United States Government incurs no responsibility or any obligation whatsoever. The fact that the Government may have formulated or in any way supplied the said drawings, specifications, or other data, is not to be regarded by implication, or otherwise in any manner construed, as licensing the holder, or any other person or corporation; or as conveying any rights or permission to manufacture, use, or sell any patented invention that may in any way be related thereto.

This report has been authored by employees of the United States Government. Accordingly, the United States Government retains a nonexclusive royalty-free license to publish or reproduce the material contained herein, or allow others to do so, for the United States Government purposes.

This report has been reviewed by the Public Affairs Office and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nationals.

If your address has changed, please notify PL/LDB, Kirtland AFB, NM 87117-6008 to help us maintain a current mailing list.

This report has been reviewed and is approved for publication.



CHARLES A. HELMS
Project Officer

FOR THE COMMANDER



STEVEN E. LAMBERSON, Lt Col, USAF
Chief, Applied Laser Technology Br



ROBERT C. LIGDAY, Lt Col, USAF
Chief, Lasers Systems Div

DO NOT RETURN COPIES OF THIS REPORT UNLESS CONTRACTUAL OBLIGATIONS OR NOTICE ON A SPECIFIC DOCUMENT REQUIRES THAT IT BE RETURNED.

REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503				
1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE January 1992	3. REPORT TYPE AND DATES COVERED Final, 1 Jul - 31 Oct 90		
4. TITLE AND SUBTITLE SINGLET DELTA OXYGEN PUMPING OF IODINE MONOFLUORIDE IN A SUPERSONIC FLOW		5. FUNDING NUMBERS PE: 62601F PR: 3151 TA: 1Z WU: 22		
6. AUTHOR(S) C. A. Helms L. Hanko G. P. Perram		G. Hager		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Phillips Laboratory Kirtland AFB, NM 87117-6008		8. PERFORMING ORGANIZATION REPORT NUMBER PL-TR--91-1044		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)		10. SPONSORING/MONITORING AGENCY REPORT NUMBER		
11. SUPPLEMENTARY NOTES				
12a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.		12b. DISTRIBUTION CODE		
13. ABSTRACT (Maximum 200 words) The iodine monofluoride (IF) molecule is known to lase via optical pumping. This report describes the first demonstration of coupling high number densities of ground state IF ($10^{15}/\text{cm}^3$) with a high concentration of singlet delta oxygen ($10^{16}/\text{cm}^3$) in a supersonic flow. Although lasing was not achieved, a qualitative description of experimental observations is presented.				
14. SUBJECT TERMS Iodine Monofluoride, Chemical-Pumping, Visible Chemical Laser, Singlet Delta Oxygen			15. NUMBER OF PAGES 32	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT SAR	

CONTENTS

<u>Section</u>	<u>Page</u>
1.0 INTRODUCTION	1
2.0 HARDWARE	2
2.1 OVERVIEW	2
2.2 FLUORINE ATOM GENERATORS	4
2.3 SPARGER GENERATOR	4
2.4 LIQUID IODINE SOURCE	8
2.5 FLUORINE AND IODINE INJECTION	8
2.5.1 Design Decisions	8
2.5.2 Hardware Description	10
3.0 EXPERIMENTAL	15
3.1 DETECTION OF $O_2(^1\Delta)$ AND $I(^2P_{1/2})$	15
3.2 GROUND STATE IF ABSORPTION	15
3.3 IF(B) NUMBER DENSITY	15
3.3.1 Description	15
3.3.2 Calculation	17
4.0 THEORY	18
5.0 RESULTS AND DISCUSSION	20
5.1 CONFIGURATION 1	20
5.2 CONFIGURATION 2	25
5.3 HF TYPE IODINE INJECTOR	25
6.0 CONCLUSIONS	25
REFERENCES	27



Accession For	
NTIS CRA&I	<input checked="" type="checkbox"/>
DTIC TAB	<input type="checkbox"/>
Unannounced	<input type="checkbox"/>
Justification	
Distribution/	
Availability Codes	
Dist. Special	
A-1	

FIGURES

<u>Figure</u>		<u>Page</u>
1.	Side view of entire device showing relative position of sparger, cold trap, combustors, and supersonic cavity.	3
2.	Drawing of F_2/NO combustors used to generate fluorine atoms for the IF Phase II program.	5
3.	Sparger type oxygen generator used as $O_2(^1\Delta)$ source.	6
4.	End and top views of cold trap section.	7
5.	Drawing of liquid iodine source.	9
6.	Assembly drawing of Configuration 1 hardware.	11
7.	Assembly drawing of Configuration 2 hardware. Fluorine atom flow was injected into the supersonic region in this configuration.	12
8.	Drawing of fast mixing nozzle. Upper view shows injection nozzles head on. Lower view shown cross section. All dimensions are in inches.	13
9.	Side view of slit iodine injection nozzle. Double plenum is present to provide even flow across the 2-in nozzle length. All dimensions are in inches.	14
10.	Schematic illustration of IF(B) number density diagnostic.	16
11.	Proposed kinetic scheme for producing IF(B) using $O_2(^1\Delta)$ as the chemical pump source.	19
12.	Emission intensity from IF(B \rightarrow X) as a function of chlorine flowrate. Data were taken from run 9263HF3. Run conditions are listed in the figure.	22
13.	Emission intensity from IF(B \rightarrow X) with and without NO present. Data were taken from run 9263FG2. Run conditions are listed in the figure.	23
14.	Emission intensity from IF(B \rightarrow X) as a function of F atom flowrate. Data were taken from run 9230HF5. Run conditions are listed in the figure.	24

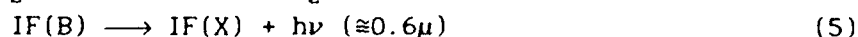
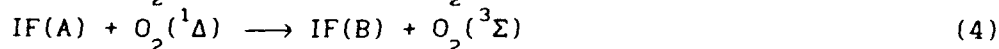
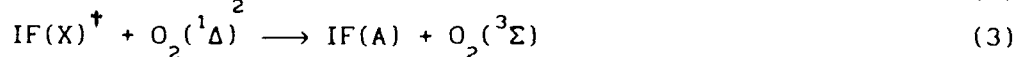
1.0 INTRODUCTION

The Department of Defense has supported research directed toward the development of chemically-pumped lasers for the last 15-20 years. This support has extended also to chemical lasers that operate in the visible region of the spectrum. Chemical lasers have been pursued as weapons systems because they are generally very efficient and have been shown to be scalable to large devices. Discussions of the strengths of visible chemical lasers have appeared elsewhere (Ref. 1).

This interest in visible chemical lasers, and specifically in an IF laser, has lead to a program at Phillips Laboratory (PL) to test the viability of a chemically-pumped IF laser. The first phase of the program demonstrated the production of ground electronic state IF (IF(X)) at a concentration of $1 \times 10^{15}/\text{cm}^3$ in a low pressure supersonic flow (Ref. 2). The topic of this technical report is the second phase of the IF program, which consisted of an effort to pump IF(X \rightarrow B) with singlet delta oxygen ($\text{O}_2(^1\Delta)$) to produce an IF laser.

Several chemical pumps for the IF(X \rightarrow B) system were considered, including active nitrogen, $\text{O}_2(^1\Delta)$, and $\text{N}_2(\text{A})$. Although active nitrogen is known to pump IF very efficiently (Ref. 3), it is not a useful energy carrier due to its high self-quenching rate (Ref. 4). Conversely, the production of large amounts of $\text{O}_2(^1\Delta)$ is a mature technology and one with which PL has much experience. Therefore $\text{O}_2(^1\Delta)$ was chosen as the pump source for IF in this work.

The principal reactions believed to be responsible for the generation of IF and subsequent chemical pumping by $\text{O}_2(^1\Delta)$ are as follows (Ref. 5). In these equations, IF(X)^\dagger represents vibrationally excited IF(X) with $v'' \geq 9$.



The second phase of the IF program was considered a high risk, high payoff project. The effort was given limited budget and manpower resources. The philosophy employed was to bring together what was believed to be the necessary species to produce high concentrations of IF(B), and then see what happened. Probably the most difficult engineering obstacle to overcome was the problem of mixing the various gas streams (O_2 , F, and I_2) on the time scale of IF(B \rightarrow X) radiative lifetime, which is 7 μ s. If quenching of IF(B) competes with radiative decay, the lifetime may be much shorter. This mixing requirement demands state-of-the-art mixing efficiency even under the best conditions.

The IF program was very hardware intensive. Although there has been considerable interest in an $O_2(^1\Delta)$ pumped IF laser, it had not been attempted on this scale before because of the budget and facility required to generate sufficiently large concentrations of $O_2(^1\Delta)$ and IF(X) in the same device. Since these hardware buildups are protracted and expensive, only a very limited number of engineering concepts can be tested in the laboratory. This process dictates the pace of the experimental work. For example, after a new design is carried through to construction and installation, its performance is evaluated experimentally. If hardware performance does not meet expectations, the evaluation period may be rather short. This was the case with every hardware configuration in Phase II of the IF program. Therefore, only qualitative or semi-quantitative observations were required to determine that the hardware built for this program was not capable of meeting its design goals.

2.0 HARDWARE

2.1 OVERVIEW

An earlier report (Ref. 6) gave a detailed account of the device (flowtube) used for the IF Phase I work. A modified version of the same device was used for the phase 2 work. This section describes each modification to the original device. A side view of the reconfigured device is shown in Figure 1.

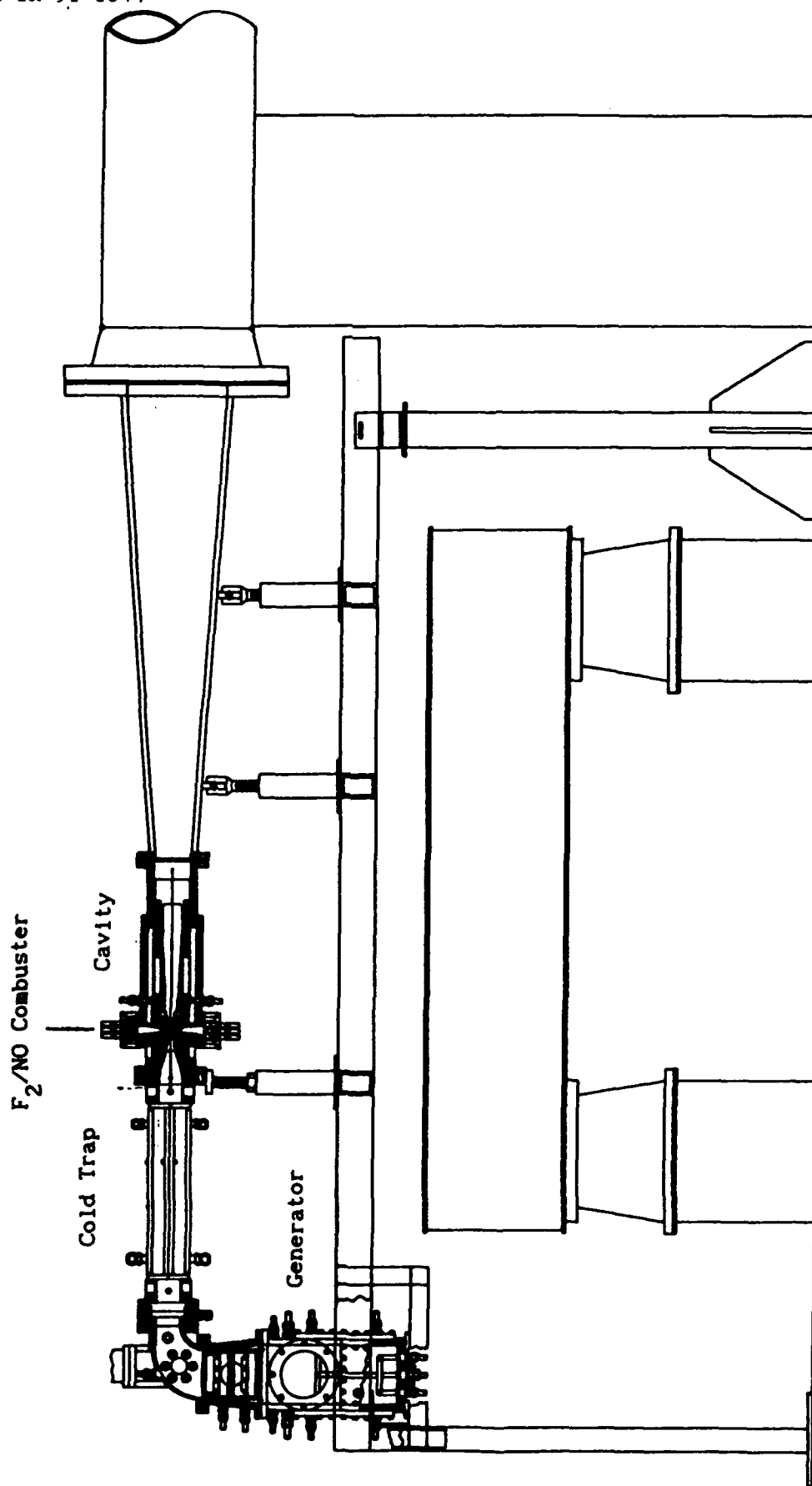


Figure 1. Side view of entire device showing relative position of sparger, cold trap, combustors, and supersonic cavity.

2.2 FLUORINE ATOM GENERATORS

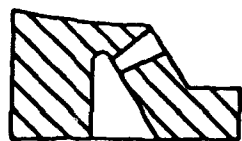
The flowtube used only one NO/F₂ combustor to generate F atoms for the Phase I work. Since F atoms were to be injected into another flow in the Phase II work, two F atom generating combustors were required to maintain a symmetric injection pattern. The internal geometry of the new combustors (volume, length, etc.) was kept very close to the original design (Fig. 2). To obtain adequate mixing of the F atoms into the oxygen flow, the combustor exit flow was forced through fifteen 0.079-in diameter holes, which were tapered out to a diameter of 0.138 in at the exit. This modification was necessary for mixing, but increased the risk of significant F atom recombination.

2.3 SPARGER GENERATOR

The generation of O₂(¹Δ) in Chemical Oxygen-Iodine Laser (COIL) devices is accomplished by reacting chlorine gas with a solution of basic hydrogen peroxide (BHP) (Ref. 7). This oxygen generation scheme may be engineered in several ways, one of which is known as a sparger generator. In this type of generator, chlorine gas is bubbled through a BHP solution that is maintained at 253-263 K. The sparger generator used on the flowtube was designed to hold about 3 l of BHP (Fig. 3). Thermal control was maintained by flowing liquid nitrogen through stainless steel heat exchange plates bolted onto the outside of the generator. The entire generator assembly was constructed of Nickel-200. Chlorine was introduced through small holes in a ring made from 0.5-in-dia stainless steel tubing that was maintained 1 to 2 in below the BHP liquid level. Helium was flowed through a second (lower) sparger ring to aid in transporting the electronically excited oxygen through the device. The BHP was prevented from leaving the generator by a set of Teflon[®] baffle plates. This arrangement is known as a liquid separator and is commonly used in COILs.

Since the oxygen generator chemicals are water based, a large amount of water vapor (several torr) is liberated from the BHP as it is held under vacuum. Water vapor has been shown (Ref. 8) to greatly reduce the power obtainable from COILs if it is allowed to reach the cavity. Cold traps are therefore commonly employed to remove the water vapor before it reaches the cavity. Figure 4 shows the configuration of the cold trap used on the flowtube.

CONFIGURATION 2



CONFIGURATION 1

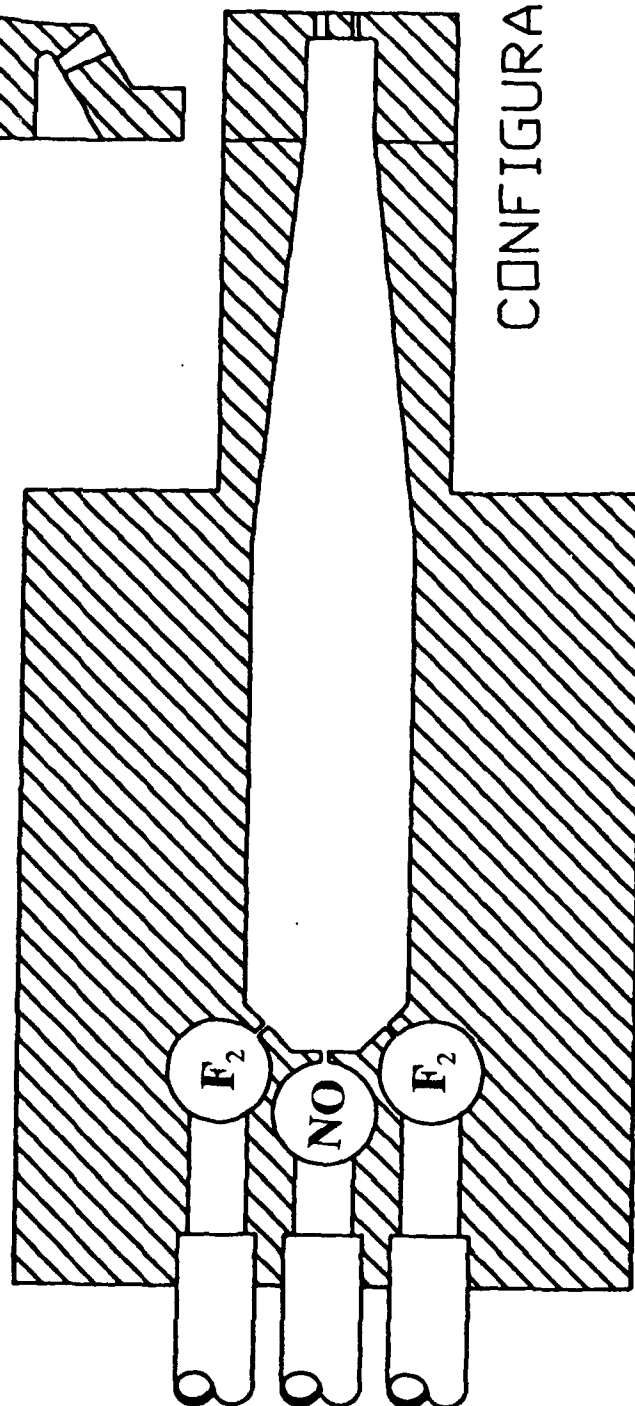


Figure 2. Drawing of F_2/NO combustors used to generate fluorine atoms for the IF Phase II program. Combustor is shown with the Configuration 1 nozzle. The Configuration 2 nozzle is also shown.

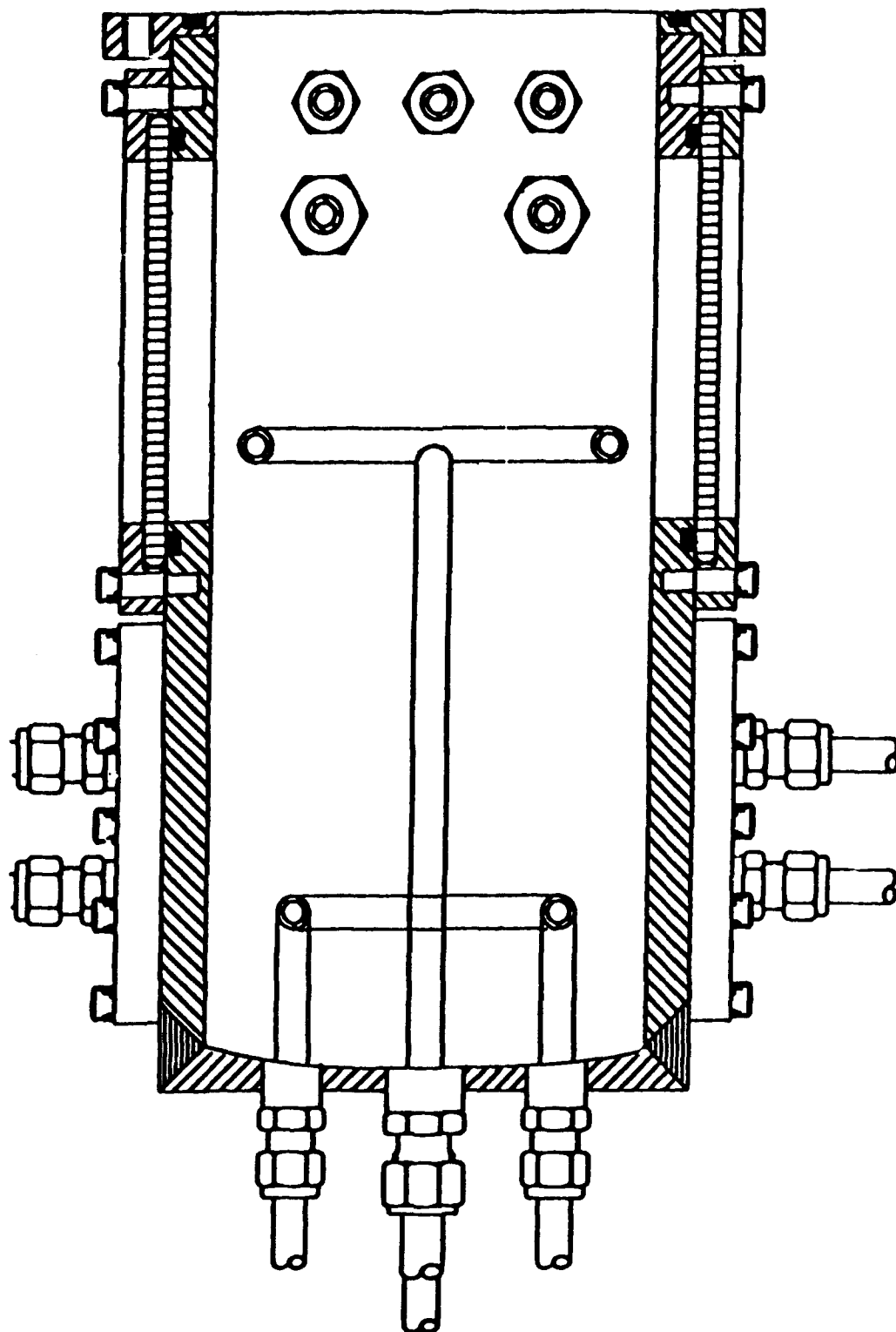


Figure 3. Sparger type oxygen generator used as $O_2(^1\Delta)$ source.

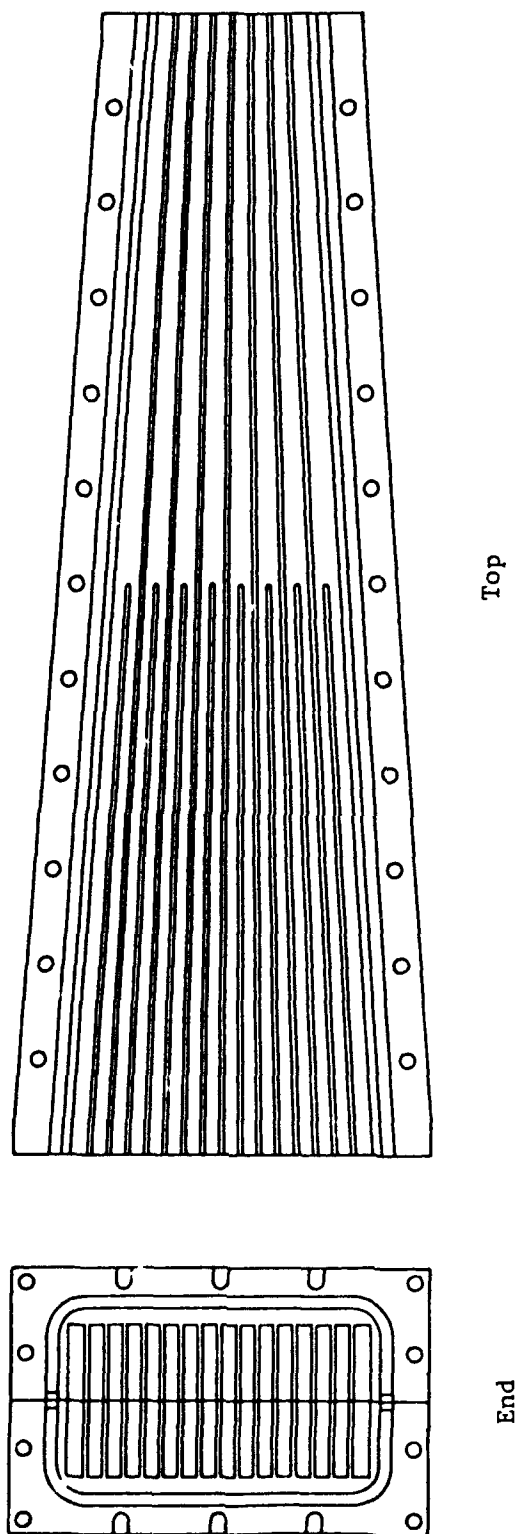


Figure 4. End and top views of cold trap section.

2.4 LIQUID IODINE SOURCE

The iodine supply to the flowtube was changed from a solid iodine bed to a liquid iodine source (Fig. 5). Diluent gas (usually helium) was bubbled through the liquid iodine and transported to the device through heated pipes. The iodine was heated to 120-140°C, depending on the desired iodine flowrate. The vessel that contained the iodine was made of hastelloy-C, an iodine resistant steel alloy.

2.5 FLUORINE AND IODINE INJECTION

2.5.1 Design Decisions

There were two critical design problems encountered in planning the $O_2(^1\Delta)$ pumped IF experiment. The first was the order of introduction of the three gas streams, $O_2(^1\Delta)$, F, and I_2 . Since the oxygen generator effluent constituted the bulk of the flow through the device, all hardware concepts involved injecting the iodine and fluorine flows into the generator flow. Thus, the only decision was whether to inject F atoms first or I_2 first. If I_2 were injected first, the I_2 might be dissociated by the $O_2(^1\Delta)$ (as occurs in COILs) before it was exposed to fluorine atoms. If the stream containing F atoms were injected first, the fluorine would have more time to recombine or react before it was exposed to I_2 . The second critical issue was mixing. Assuming that the first two gas streams were thoroughly mixed (F or I_2 with $O_2(^1\Delta)$), the third gas had to be injected such that mixing occurred fast enough to compete with vibrational quenching and radiative relaxation (see Section 4.0). The required mixing time is probably $<10 \mu s$. This mixing time is difficult (if not impossible) to achieve due in part to the high molecular weight of I_2 .

Both of these problems were dealt with in hardware design by testing several different injection schemes. The order of injection was addressed by designing two sets of hardware that allowed the user to inject F and I_2 in either order. However, other than the Configuration 2 hardware described below, iodine was always injected last, in the supersonic expansion region of the flow. The mixing problem was more difficult. A complete fluid dynamic modeling of the mixing situation was well beyond the scope of the available

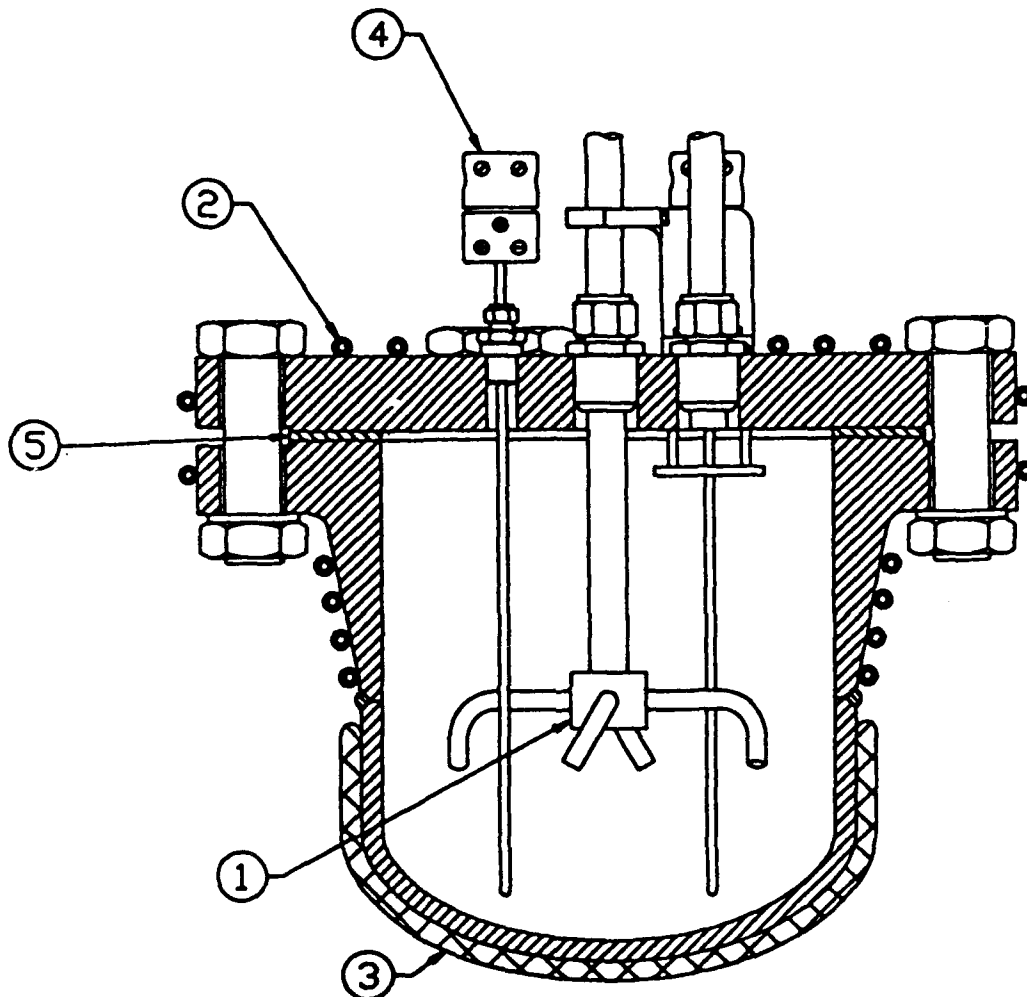


Figure 5. Drawing of liquid iodine source. Main features include; (1) helium injection rake; (2) rope heaters; (3) flexible heating mantle; (4) Type T thermocouple (two shown); (5) Teflon gasket seal.

time and budget. Therefore, for the initial hardware configuration, an educated guess was made to determine injection hole sizes, spacing, and location for the third gas stream to be introduced to the flow. The following injection schemes are listed in chronological order. None of the iodine injectors described below was actively heated other than by the gas flow.

2.5.2 Hardware Description

The first set of hardware used is shown in Figures 6 and 7. The Configuration 1 hardware allowed the fluorine atoms to be injected in the subsonic region and the iodine to be injected into the supersonic expansion. The Configuration 2 hardware reversed the order of injection. This hardware was designed in-house and fabricated at the PL machine shop. Results from this hardware indicated that much smaller iodine injection holes were needed.

The next injection scheme was taken directly from Hydrogen Fluoride (HF) laser technology. Because very extensive fluid dynamic modeling had been done for HF lasers to achieve optimum mixing, their resulting hardware design was incorporated into the IF hardware. Aluminum versions of the HF laser injection hardware were obtained (Fig. 8). Since aluminum is incompatible with iodine, stainless steel copies were fabricated for use on the flowtube.

There are major differences between the HF and IF mixing situations, including plenum pressures, molecular weights, and flowrates. These important differences render the validity of copying the HF hardware suspect at best. Nonetheless, this approach yielded a mixing situation that the fluid mechanics advisors to the IF program believed would work as well as any other options available.

Another iodine injection scheme was designed and is shown in Figure 9. The idea behind this injector was to minimize mixing of iodine into the main flow by injecting a thin sheet of iodine. The slow diffusion limited mixing that would result from this injector was predicted to be easier to model and measure. The injection slit was 5 cm wide and 0.0094 in high, and was fed from both ends with iodine.

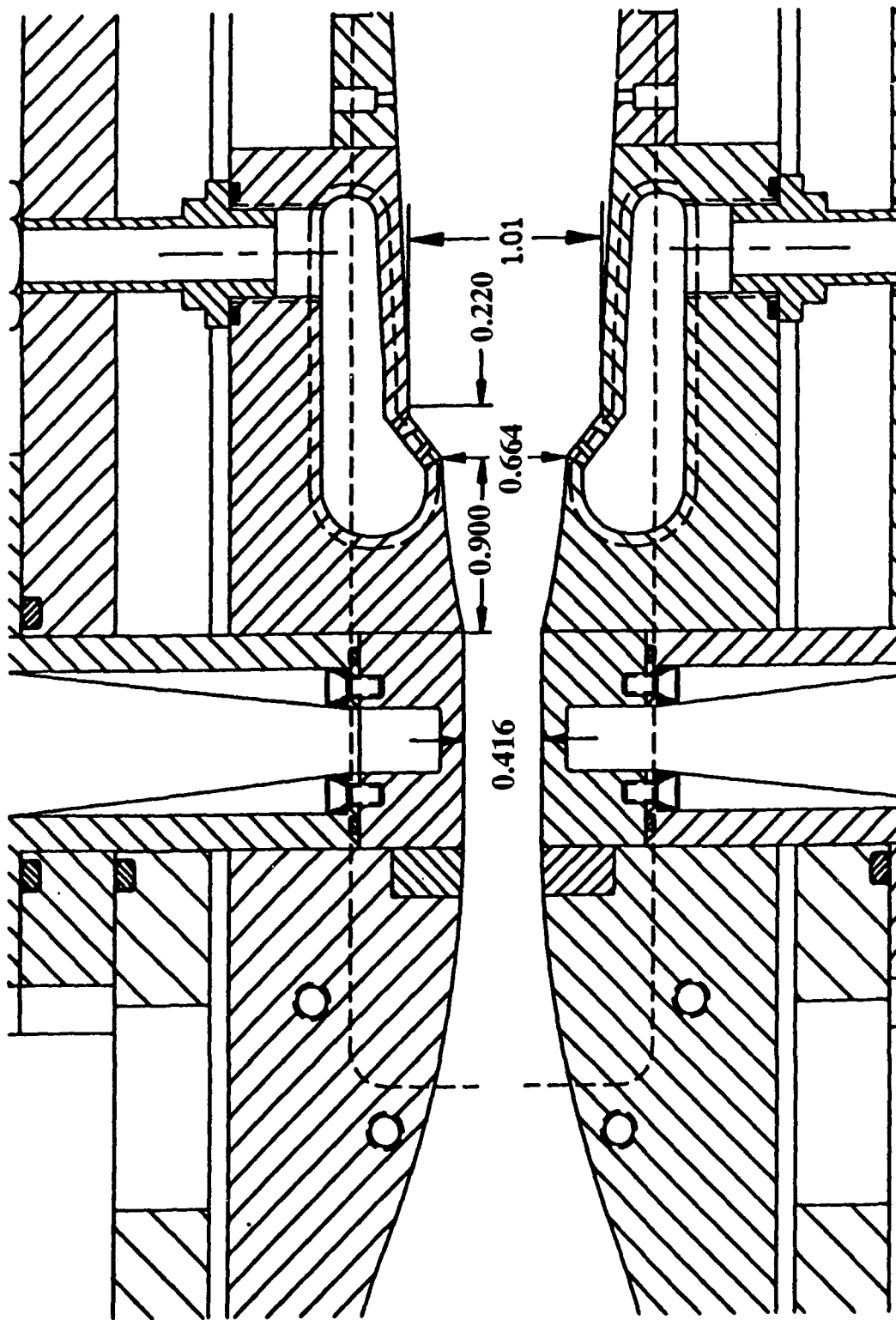


Figure 6. Assembly drawing of Configuration 1 hardware. Iodine delivery is in the supersonic region in this scheme.

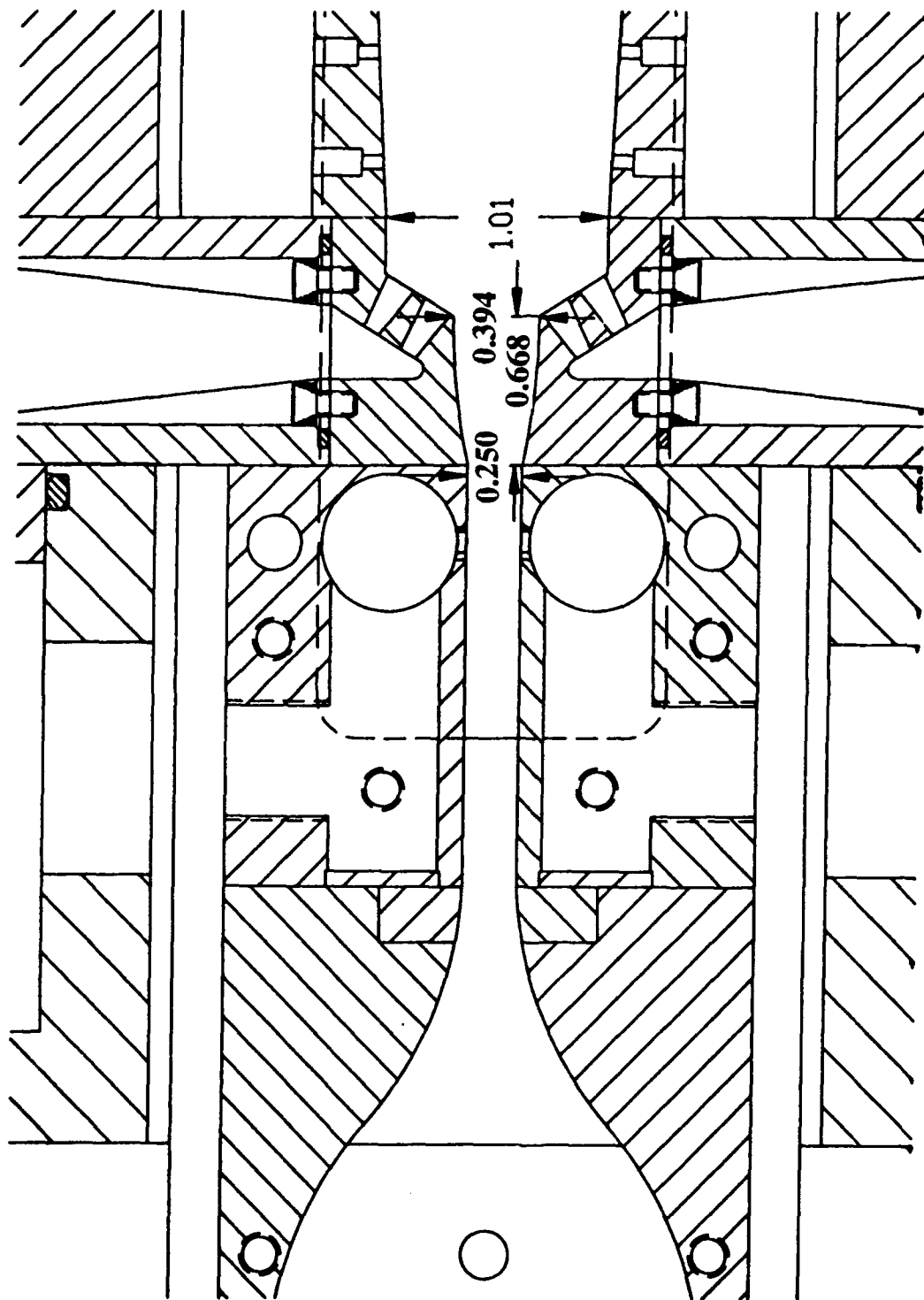


Figure 7. Assembly drawing of Configuration 2 hardware. Fluorine atom flow was injected into the supersonic region in this configuration.

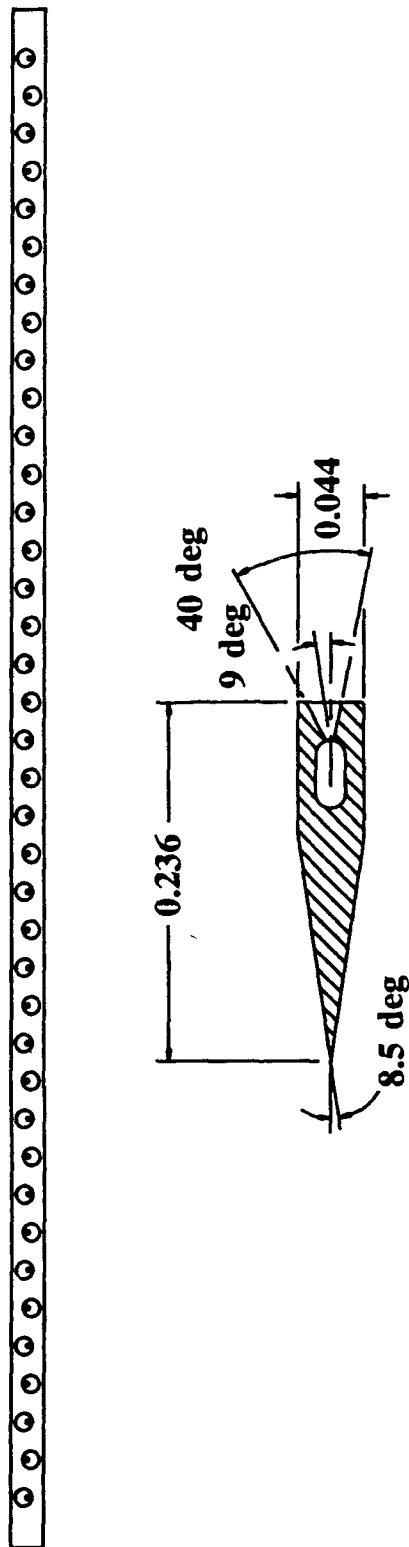


Figure 8. Drawing of fast mixing nozzle. Upper view shows injection nozzles head on. Lower view shows cross section. All dimensions are in inches.

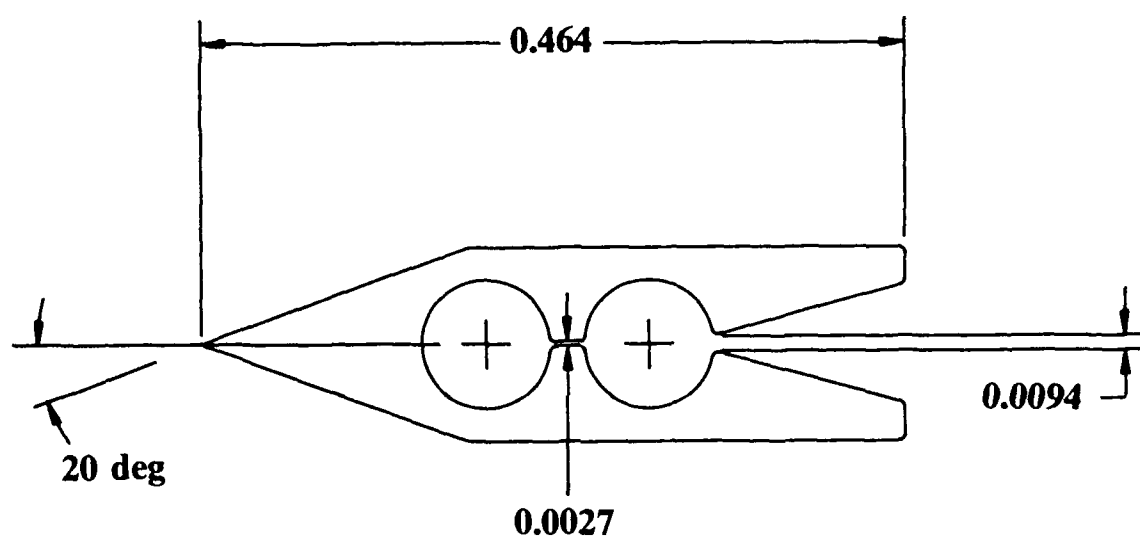


Figure 9. Side view of slit iodine injection nozzle. Double plenum is present to provide even flow across the 2-in nozzle length. All dimensions are in inches.

To test some optical diagnostics while waiting for hardware, a few iodine injection nozzles were made on site. These nozzles were steel tubing of 0.125- and 0.25-in diameter with about twenty 0.013-in-dia holes, the smallest drill bits on hand. The drilling was done by a technician in about an hour. Iodine was fed from only one end; the other end was crimped and welded closed.

3.0 EXPERIMENTAL

3.1 DETECTION OF $O_2(^1\Delta)$ AND $I(^2P_{1/2})$

The performance of the sparger generator was monitored by measuring the emission from $O_2(^1\Delta)$ at $\lambda = 1.27 \mu m$. An Applied Optics Corporation model 430L intrinsic germanium detector (Wang) was used to monitor the $1.27 \mu m$ radiation. The calibration and proper use of this detector has been described in detail elsewhere.* Emission was recorded immediately upstream and downstream of the cold trap section. This diagnostic was essential to establish proper operation of the sparger generator. A Wang detector was also used to measure $I(^2P_{1/2})$ emission in the cavity at $1.315 \mu m$.

3.2 GROUND STATE IF ABSORPTION

Ground state iodine monofluoride ($IF(X^1\Sigma^+)$) was measured by absorption with a Coherent model 699-29 ring dye laser pumped by a Coherent model CR3000K krypton ion laser. Output from the ring laser was sufficiently narrow (≈ 20 MHz) to resolve individual rotational lines. This technique was also used for the IF Phase I effort and was described in an earlier report (Ref. 2).

3.3 IF(B) NUMBER DENSITY

3.3.1 Description

A quantitative measure of the number density of $IF(B^3\Pi(O^+))$ in the flowtube cavity was made by directly observing the $IF(B, v' = 0 \rightarrow X, v'' = 4)$ emission at 603 nm. A schematic of the experimental arrangement is shown in Figure 10.

* Keating, P., Hanko, L., Helms, C.A., Perram G.P., "Absolute Detection of $O_2(^1\Delta)$ Concentrations," WL-TR-90-85, Weapons Laboratory, Kirtland AFB, NM 87117, to be published.

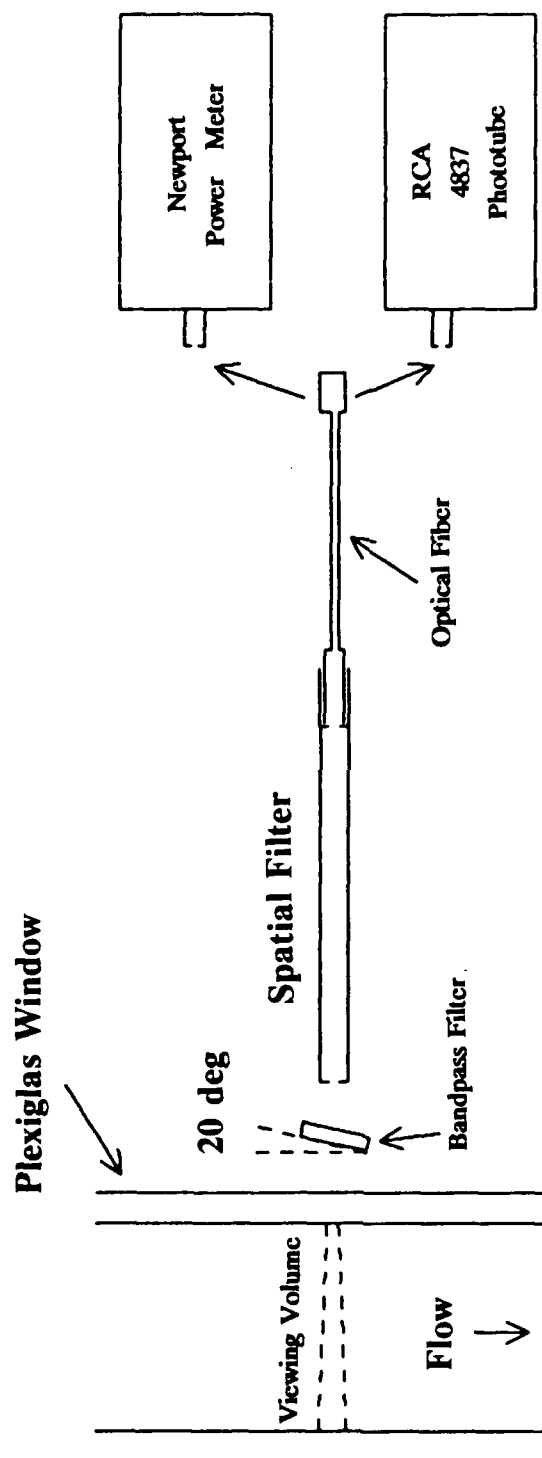


Figure 10. Schematic illustration of IF(B) number density diagnostic.

A 600-nm narrow bandpass filter (Corion G522-6200 10 nm FWHM) was rotated 20 deg from perpendicular to the optical axis to center the filter transmission at 603.4 nm. This permitted only the $IF(B, v' = 0 \rightarrow X, v'' = 4)$ band to pass through the optical train. The viewing volume of the emission was defined by a spatial filter that has been described elsewhere (Ref. 9). Briefly, the spatial filter is a black anodized aluminum tube with entrance and exit apertures adjustable down to 1 mm and with internal baffles to block reflected light. Light transmitted through the spatial filter was coupled into an optical fiber which led to some type of detector. The system was calibrated by alternately permitting light exiting the fiber to impinge on an NIST traceable Newport Research Corporation model 830 power meter and an RCA model 4837 photomultiplier tube. The phototube was supplied with a 900-V negative bias. An incandescent bulb was used as the light source to cross-calibrate the photomultiplier tube to the power meter with the bandpass filter in place. This procedure was performed several times to check to the repeatability of inserting the fiber into its holder in front of the two detectors. The following discussion describes how $IF(B)$ number density was derived from photomultiplier output current.

3.3.2 Calculation

The photomultiplier tube response was determined to be 1 nA output per picowatt of optical input at 603 nm. The volume of the gas flow region was 0.35 cm^3 , which was defined by the spatial filter. Since light was emitted isotropically from this volume, the solid angle of light collected by the spatial filter had to be considered. The geometry used in these experiments collected 8×10^{-7} of the total light emitted. Several optical losses also had to be accounted for, such as the bandpass filter (Transmission = 35%), the plexiglas device window ($T = 80\%$), and the optical fiber ($T = 50\%$).

The total number density of $IF(B)$ is related to the number density of $IF(B, v' = 0)$ by

$$[IF(B)] = \frac{[IF(B, v' = 0)]}{F_{vib}} \quad (6)$$

where F_{vib} ($= 0.65$ @ 600K) is the fraction of $\text{IF}(\text{B})$ in the $v' = 0$ vibrational level. The number density of $\text{IF}(\text{B}, v' = 0)$ is related to the rate of spontaneous emission by the first order radiative decay of $\text{IF}(\text{B})$, given by

$$-\frac{d[\text{IF}(\text{B}, v' = 0)]}{dt} = k_r [\text{IF}(\text{B}, v' = 0)] = \frac{\text{photons emitted}}{\text{cm}^3 \text{ sec}} \quad (7)$$

where k_r ($= 1.7 \times 10^5$) is the radiative rate constant for $\text{IF}(\text{B} \rightarrow \text{X})$. In turn, the rate of emission of photons due to $\text{IF}(\text{B}, v' = 0 \rightarrow \text{X}, v'' = 4)$ can be determined from

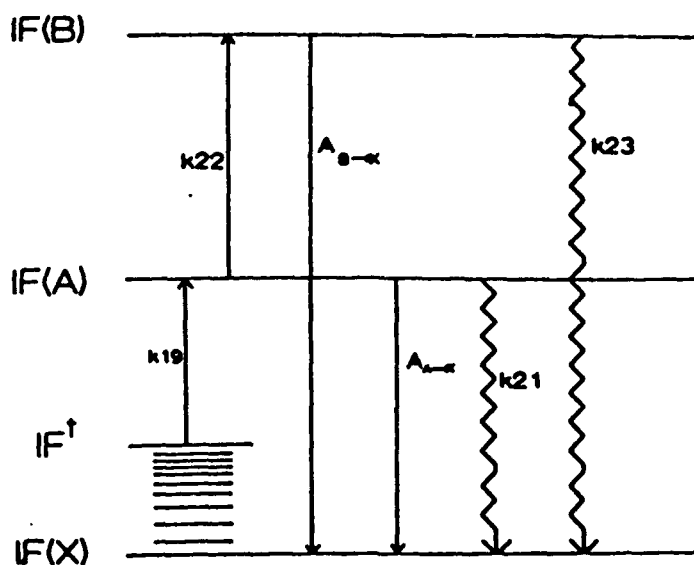
$$\frac{\text{photons emitted}}{\text{cm}^3 \text{ sec}} = \frac{\text{photons detected}}{\text{sec}} \cdot F_{\text{ang}} \cdot T \cdot V \cdot \left[\frac{q_{0 \rightarrow 4}}{\sum_i q_{0 \rightarrow i}} \right]^{-1} \quad (8)$$

where F_{ang} ($= 8 \times 10^{-7}$) is the fractional part of the total emission collected by the spatial filter, T ($= 0.14$) is the optical throughput, V ($= 0.35 \text{ cm}^3$) is the active viewing region, and $q_{0 \rightarrow i}$ is the Frank-Condon factor for $v' = 0$ to $v'' = i$. Finally, the number of photons detected per second is related to the phototube output current by

$$\frac{\text{photons detected}}{\text{sec}} = \text{phototube current} \cdot \frac{\text{optical power}}{\text{tube current}} \cdot \frac{\text{photons detected}}{\text{watt sec}} \quad (9)$$

4.0 THEORY

As mentioned in the introduction, $\text{IF}(\text{X})^\dagger$ is believed to play an important role in chemically pumping $\text{IF}(\text{X} \rightarrow \text{B})$ using $\text{O}_2(^1\Delta)$ as the energy donor. An IF pump mechanism reflecting this has been proposed by Whitefield, et al, in an earlier paper (Ref. 5), and is shown in Figure 11. Davis, et al, (Ref. 10) found that $\text{O}_2(^1\Delta)$ was much more efficient at pumping IF when $\text{IF}(\text{X})$ was produced by the $\text{F} + \text{I}_2$ reaction (reaction [1] shown above). This production scheme for $\text{IF}(\text{X})$ is unique because the $\text{IF}(\text{X})$ is born with a strongly inverted vibrational distribution, with little $v'' < 6$ (Refs. 10 and 11). This is significant because $\text{O}_2(^1\Delta)$ only has sufficient energy to pump $\text{IF}(\text{X} \rightarrow \text{A})$ for $v'' \geq 8$.



(a) IF level diagram.

$$[IF(B)] = \frac{k_{19}k_{22}[IF^{\dagger}][O_2(^1\Delta)]^2}{(A_{g \rightarrow x} + k_{23}[M])(A_{x \rightarrow g} + k_{21}[M] + k_{22}[O_2(^1\Delta)])}$$

(b) Expression for IF(B) number density using proposed mechanism and the steady-state approximation.

Figure 11. Proposed kinetic scheme for producing IF(B) using O₂(¹Δ) as the chemical pump source.

The $F + I_2$ reaction was chosen for the IF program at PL to take advantage of this nascent vibrational inversion. Effective use of the initial vibrational energy requires a collision between $IF(X)^{\dagger}$ and $O_2(^1\Delta)$ on at least the same time scale as ground state vibrational relaxation. Mixing an iodine stream into a F/O_2 stream on this time scale appears to be beyond current hardware capabilities.

Reaction (2) requires a collision between $O_2(^1\Delta)$ and $IF(A)$. This must compete with collisions of $IF(A)$ with quenchers in the flow and with radiative losses. Finally, to produce high concentrations of $IF(B)$ the rate of production of $IF(B)$ must be comparable to the radiative decay rate ($7 \mu s$). Furthermore, although most species present in the flow are known to not quench $IF(B)$ efficiently, the quenching rate of $IF(B)$ due to nitrosyl fluoride (FNO) is not known. If this or other potential quenching reactions are fast, the measured lifetime of $IF(B)$ in the flow may be significantly shorter than $7 \mu s$. All of these factors together indicate that demonstrating an inversion on $IF(B \rightarrow X)$ through collisions with $O_2(^1\Delta)$ is a difficult undertaking that depends heavily on how rapidly the three gas flows can be mixed.

5.0 RESULTS AND DISCUSSION

5.1 CONFIGURATION 1

Initial experiments involved introducing all reactive gases to get an impression of how bright the mixing region would be. The $IF(B \rightarrow X)$ emission is clearly visible with only fluorine atoms and iodine present. The $IF(B \rightarrow X)$ emission observed in the absence of $O_2(^1\Delta)$ is believed to be caused by $I(^2P_{1/2})$ pumping $IF(X \rightarrow B)$. The $I(^2P_{1/2})$ is thought to be a minor product of the $F + I_2$ reaction. The overall emission became much brighter upon the addition of chlorine and was by far the brightest at the interface between the iodine injection jets and the surrounding flow. This is not surprising since the interface is where IF is produced and is in the presence of $O_2(^1\Delta)$. Obtaining an accurate $IF(B)$ number density measurement from this bright region was not possible because diagnostic beams or viewing volumes only provide an average number density over the probed volume.

Several tests were conducted in an attempt to corroborate or refute the proposed model for IF excitation. The principal diagnostic for these tests was an EG&G PAR model 1421 optical multichannel analyzer (OMA) detector exposed to the entire emission region in the cavity. The monochromator (without the exit slit) was tuned to allow roughly 575- to 625-nm light to reach the OMA. Conclusions were drawn based on relative emission intensities under different flow conditions.

One such test was to vary the chlorine flowrate to evaluate the effectiveness of $O_2(^1\Delta)$ as a chemical pump for IF under our operating conditions. As shown in Figure 12, the IF(B-X) emission intensity increased dramatically as the chlorine was turned on. Further increases in chlorine flowrate did not, however, produce corresponding increases in the emission intensity.

Another test was performed to verify the effectiveness of using F atoms in favor of molecular fluorine to convert I_2 to IF(X). Much more IF(B) emission was expected using fluorine atoms both because F reacts more rapidly with I_2 than does F_2 , and $F + I_2$ produces vibrationally excited IF(X), which may aid in the pumping process. A great enhancement in emission was indeed observed when the nitric oxide flow was on (Fig. 13). However, ramping the F atom flow by ramping the NO and F_2 flows did not produce continuously increasing emission (Fig. 14).

Thirty-five vibrational bands were assigned to IF(B-X) from 500 to 700 nm, yielding a vibrational temperature of roughly 1200 K, in good agreement with IF Phase I results (Ref. 2).

The presence of $O_2(^1\Delta)$ in the cavity section was confirmed by monitoring $I(^2P_{1/2})$ emission in the cavity as a function of downstream position. Also, $I_2(B-X)$ emission was monitored in the cavity, and decreased downstream as expected due to reaction with F and dissociation of any excess by $O_2(^1\Delta)$.

Although these results are consistent with known properties of this system and the proposed pump mechanism, no quantitative measurements were obtained with this hardware. The important conclusion was that this hardware was not capable of introducing the iodine stream into the $O_2(^1\Delta)$ /F stream fast enough

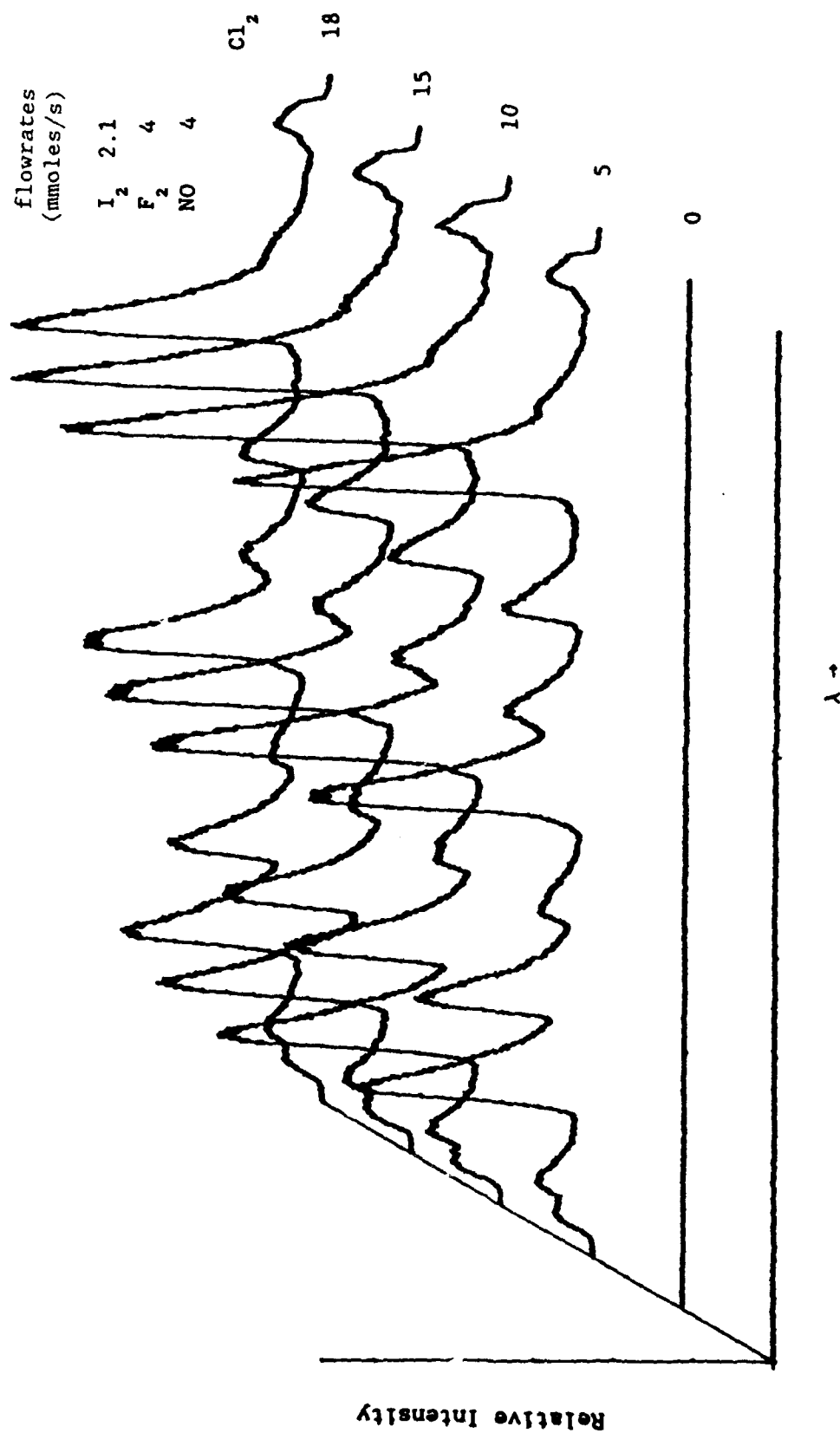


Figure 12. Emission intensity from $IF(B \rightarrow X)$ as a function of chlorine flowrate. Data were taken from run 9263HF3. Run conditions are listed in the figure.

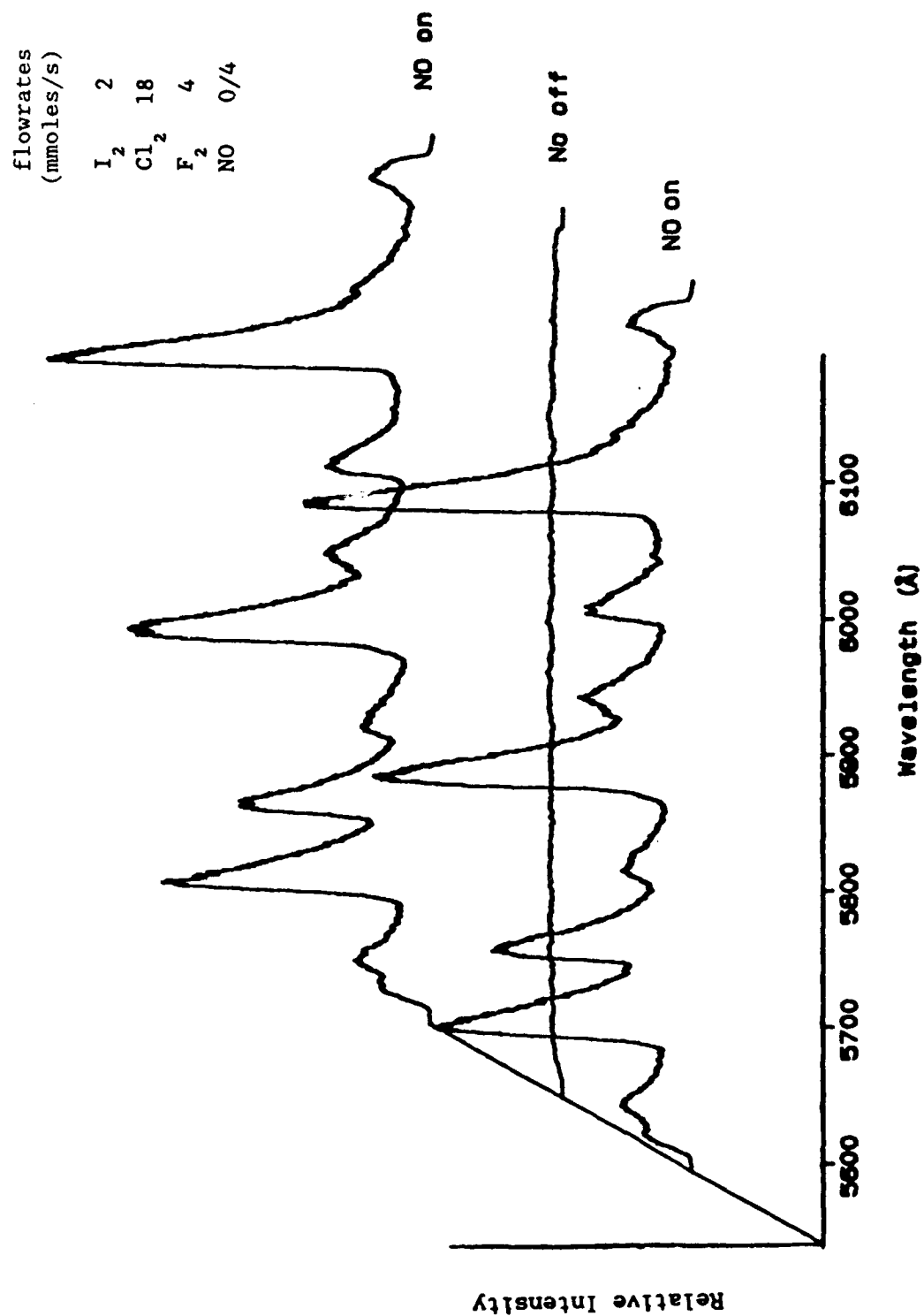


Figure 13. Emission intensity from IF(B→X) with and without NO present. Data were taken from run 9263FG2. Run conditions are listed in the figure.

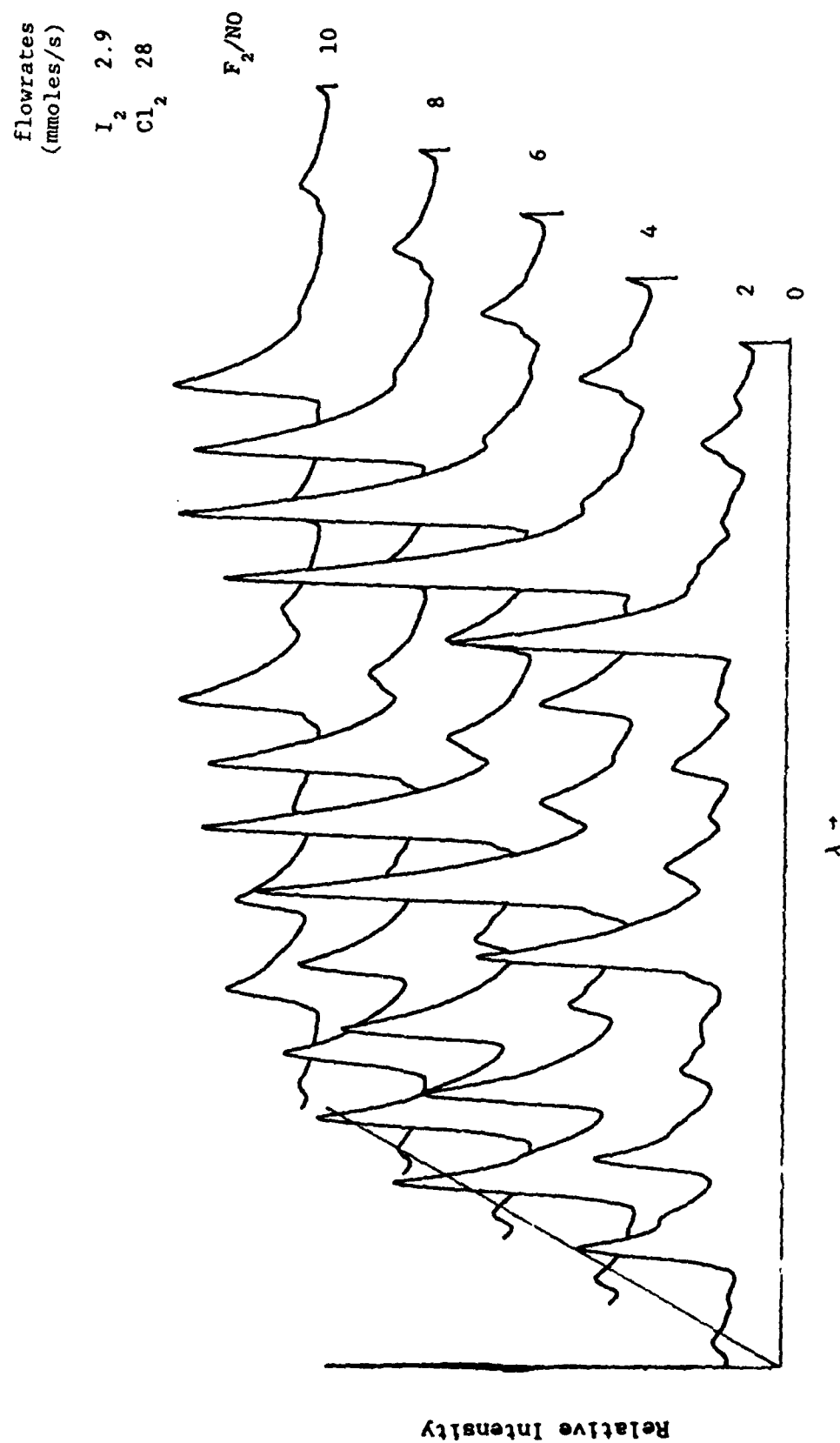


Figure 14. Emission intensity from IF(B-X) as a function of F atom flowrate. Data were taken from run 9230HF5. Run conditions are listed in the figure.

to produce large number densities of IF(B) in a sufficiently large volume to be useful for diagnostic measurements.

5.2 CONFIGURATION 2

Configuration 2 hardware (iodine injection upstream of fluorine) was not examined nearly as thoroughly as Configuration 1. Testing of this hardware consisted of a check of the intensity of IF(B) emission and a visual evaluation of mixing efficiency. The individual F/FNO jets entering the supersonic flow were still easily distinguishable, again indicative of poor mixing. Since this order of injection did not appear to be clearly superior in terms of mixing quality and dissociation of I_2 by $O_2(^1\Delta)$ prior to F injection was a concern in this hardware scheme, all subsequent hardware designs allowed for downstream iodine injection only.

5.3 HF TYPE IODINE INJECTOR

Initial experiments using the "fast mixing nozzle" fashioned after HF/DF injectors again produced poor results. Even with injection holes as small as 0.007 in, individual jets were observed and no homogeneous bright region existed with which to obtain a good IF(B) number density measurement. Other problems plagued this setup, such as low iodine flow due to the very small effective iodine orifice, and plugging of the small holes with iodine.

An IF(B) number density of about $10^9/\text{cm}^3$ was measured, but again this value is an average over the viewing volume of the spatial filter, and is not a good indication of the IF(B) number density in the bright interface region where the gases mix.

6.0 CONCLUSIONS

Mixing limitations rendered the experimental results obtained in this work nearly uninterpretable. Until these mixing problems are solved, it will be very difficult to evaluate the potential of $O_2(^1\Delta)$ pumped IF as a viable chemical laser. However, one conclusion is that making an $O_2(^1\Delta)$ -pumped IF laser is not as simple as getting large concentrations of the pump and lasing molecules into the same flow.

Although the kinetics and physical properties of IF and O_2 have been shown to be favorable for a laser system, there are issues that may prove fatal to a working system. The quenching rate of IF(B) by FNO, a major product of F atom generation, could be high enough to prevent efficient operation of an IF laser. Mixing iodine into the flow fast enough to use the nascent vibrational energy may not be possible.

It should be noted that the IF program was ended partly due to time limitations. The failure here to produce a high concentration of IF(B) in a useful volume does not mean that it cannot be done. More exotic mixing schemes may be able to use the excess vibrational energy available as proposed in the model. Also, other pump sources may turn out to be more useful than $O_2(^1\Delta)$.

REFERENCES

1. Davis, S.J., "Prospects for Visible Chemical Lasers," Procs. SPIE 540 p. 188, 1985.
2. Helms, C.A., Hanco, L., Healey, K., Hager, G., and Perram, G.P., "Generation of High Number Densities of Iodine Monofluoride in a Supersonic Flow," J. Appl. Phys. 66, p. 6093, 1989.
3. Piper, L.G., Marinelli, W.J., Rawlins, W.T., and Green, B.D., "The Excitation of $IF(B^3\Pi_0^+)$ by $N_2(A^3\Sigma_u^+)$," J. Chem. Phys. 83 p. 5602, 1985.
4. Clark, W.G., Setser, D. W., "Energy Transfer Reactions of $N_2(A^3\Sigma^+)$. 5. Quenching by Hydrogen Halides, Methyl Halides, and Other Molecules," J. Phys. Chem. 84 p. 2225, 1980.
5. Whitefield, P.D., Shea, R.F, and Davis, S.J., "Singlet molecular oxygen pumping of $IF\ B^3\Pi(0^+)$," J. Chem. Phys. 78 p. 6793, 1983.
6. Helms, C.A., Hanco, L., Miller, J., Healey, K., Hager, G., and Perram, G.P., "Generation of Iodine Monofluoride in a Supersonic Multipurpose Flowtube Facility," AFWL-TR-89-29, Air Force Weapons Laboratory, Kirtland AFB, NM 87117, August 1989.
7. Avizonis, P., et al., "Report to the American Physics Society of the Study Group on Science and Technology of Directed Energy Weapons," Rev. Mod. Phys. 59 p. S43, 1987.
8. Zagidullin, M.V., Igoshin, V.I., Katulin, V.A., and Kupriyanov, N.L., "Numerical Modeling of a Chemical Oxygen-Iodine Laser," Sov. J. Quantum Electron. 12 p. 1235, 1982.
9. Johnson, D.E., "Parametric Studies of I_2 Dissociation and I^* Production in Supersonic Slit Nozzles," AFWL-TR-86-116, Air Force Weapons Laboratory, Kirtland AFB, NM 87117, April 1987.
10. Davis, S.J., Woodward, A.M., Piper, L.G., and Marinelli, W.J., "Chemical Pump Sources for $IF(B)$," PSI-2000/TR-689, Physical Sciences Inc., Research Park, P.O. Box 3100, Andover, MA 01810, June 1987.
11. Girard, B., Billy, N., Bouedard, G. and Vigue, J. "LIF Study of the $I + F \rightarrow IF + I$ Reaction: Population of the $v'' = 0$ Level of IF ," Chem. Phys. Lett. 136 p. 101, 1987.